Correlated neutron emission in fission

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Abstract. We have implemented a Monte-Carlo simulation of the fission fragments statistical decay by sequential neutron emission. Within this approach, we calculate both the center-of-mass and laboratory prompt neutron energy spectra, the prompt neutron multiplicity distribution P(v), and the average total number of emitted neutrons as a function of the mass of the fission fragment $\bar{v}(A)$. Two assumptions for partitioning the total available excitation energy among the light and heavy fragments are considered. Preliminary results are reported for the neutron-induced fission of 235 U (at 0.53 MeV neutron energy) and for the spontaneous fission of 252 Cf.

INTRODUCTION

In this work, we extend the Los Alamos model [1] by implementing a Monte-Carlo simulation of the statistical decay (Weisskopf-Ewing) of the fission fragments (FF) by sequential neutron emission. This approach leads to a much more detailed picture of the decay process and various physical quantities can then be assessed: the center-of-mass and laboratory prompt neutron energy spectrum $N(\varepsilon_n)$, the prompt neutron multiplicity distribution P(v), the average number of emitted neutrons as a function of the FF $\bar{v}(A,Z)$, and all possible neutron-neutron correlations.

The layout of this paper is as follows: first, our theoretical and modeling approach are introduced. Numerical results on both the spontaneous fission of 252 Cf and neutron-induced (e_n =0.53 MeV) fission of 235 U are then presented. Finally, a discussion of these preliminary results and open questions conclude the paper.

THEORETICAL APPROACH

Unlike in the Los Alamos model where many quantities are lumped together, our approach tries to follow in detail the statistical decay of the FF by sequential emission of individual neutrons. Many quantities and parameters, more or less known, enter as input in the calculation. We will now briefly describe the methodology used in the present work, and then go into some detail over the list of input quantities that enter in our calculations.

Methodology

A Monte Carlo approach allows to follow in detail any reaction chain and to record the result in a history-type file, which basically mimics the results of an experiment.

We first sample the FF mass and charge distributions, and pick a pair of light and heavy nuclei that will then decay by emitting zero, one or several neutrons. This decay sequence is governed by neutrons emission probabilities at different temperatures of the compound nucleus and by the energies of the emitted neutrons.

The FF mass and charge distributions is given by $Y(A,Z) = Y(A)_{exp} \times P(Z)$, where $Y(A)_{exp}$ represents an experimental pre-neutron FF mass distribution. The charge distribution P(Z) is assumed Gaussian in shape.

Of course, the particular decay path followed by this pair of nuclei depends on the available excitation energies, which can be deduced in the following manner. The total excitation energy available for the pair $(A,Z)_l$ (light), $(A,Z)_h$ (heavy) reads

$$E_T^*(A_l, A_h, Z_l, Z_h) = E_r^*(A_l, A_h, Z_l, Z_h) + B_n(A_c, Z_c) + e_n - TKE(A_l, A_h),$$
(1)

where $E_r^*(A_l, A_h, Z_l, Z_h)$ is the energy release in the fission process, which is given, in the case of binary fission, by the difference between the compound nucleus and the FF masses. $B_n(A_c, Z_c)$ and e_n are the separation and kinetic energies of the neutron inducing fission (in the case of spontaneous fission, both $B_n(A_c, Z_c)$ and e_n terms in Eq. (1) disappear). $TKE(A_l, A_h)$ is the total FF kinetic energy. In fact, TKE is not a single value but rather a distribution, assumed to be Gaussian, whose mean value and width are taken from experiment.

One of the long-standing questions about the nuclear fission process is how does the available total excitation

energy get partitioned among the light and heavy fragments. In the present study, we have considered two hypotheses for partitioning this energy:

• Partitioning so that both light and heavy fragments share the same temperature (hypothesis identical to the one made in the Los Alamos model [1]) at the instant of scission. From this condition, it follows that the initial excitation energy of a given FF is:

$$E_{l,h}^* = E_T^* \frac{1}{1 + \frac{a_{h,l}}{a_{l,h}}},\tag{2}$$

where l and h refer to the light and heavy system.

• Partitioning using the experimental $\bar{v}(A)$ to infer the initial excitation of each fragment. This condition writes as follow:

$$E_{l,h}^* = E_T^* \frac{\bar{\mathbf{v}}(A_{l,h})\langle \varepsilon \rangle_{l,h}}{\sum_{i=l,h} \bar{\mathbf{v}}(A_i)\langle \varepsilon \rangle_i},\tag{3}$$

where $\langle \varepsilon \rangle_{l,h}$ is equal to the average energy removed per emitted neutron ($\varepsilon = 1.265$ and 1.511 MeV for the n (0.53 MeV) + 235 U and 252 Cf(sf) respectively). It is the sum of the average center-of-mass energy of the emitted neutrons and of the average FF neutron separation energy.

Within the Fermi-gas model, the initial FF excitation energy $E_{l,h}^*$ is simply related to the nuclear temperature $T_{l,h}$. The probability for the FF to emit a neutron at a given kinetic energy is obtained by sampling over the Weisskopf spectrum at this particular temperature [2]:

$$\phi(A, Z, \varepsilon_n, T) = \frac{\varepsilon_n}{T_{A,Z}^2} e^{\frac{-\varepsilon_n}{T_{A,Z}}}, \tag{4}$$

where $T_{A,Z}$ is the nuclear temperature of the residual nucleus given by

$$T_{A,Z} = \sqrt{\frac{E^*(A,Z) - B_n(A,Z)}{a_{A-1,Z}}},$$
 (5)

with $a_{A,Z}$ the level density parameter of the nucleus.

The emission of a neutron of energy ε_n from the FF at the excitation energy E^* produces a residual nucleus with the excitation energy

$$E^*(A-1,Z) = E^*(A,Z) - \varepsilon_n - B_n(A,Z).$$
 (6)

The sequential neutron emission ends when the excitation energy of the residual nucleus is less than the sum of its neutron separation energy and pairing energy.

The transformation of the center-of-mass spectrum to the laboratory spectrum is done by assuming that neutrons are emitted isotropically in the center-of-mass frame of a FF. So, sampling over the angle of emission of the neutron $\theta_n \in [0, \pi]$ for each nucleus (A, Z), we infer the neutron energy in the laboratory frame, taking into account the recoil energy of the residual nucleus.

Input Parameters

The fission mass yields have been measured extensively and precisely for several nuclei and energies. In the present calculation, we sample over the pre-neutron fragments yields Y(A), i.e., before neutron evaporation, as reconstructed from the experimentally measured fission products mass distribution. In particular, we use the data by Hambsch [3] in the case of 252 Cf(sf), and the data by Schmitt [4] in the case of the neutron-induced fission (at 0.53 MeV) on 235 U.

255 fragments were used to represent the Y(A,Z) for the neutron induced $n(0.53 \text{ MeV}) + ^{235}\text{U}$ reaction. In particular, we considered 85 equispaced fragment masses (between $76 \le A \le 160$) with 3 isobars per fragment mass, around the most probable charge Z_p . In the case of spontaneous fission of ^{252}Cf , we used 315 FF between $74 \le A \le 178$ among which 105 fragment masses.

Nuclear masses are used to calculate the energy release for a given pair of FF. It is a function of both mass and charge number of complementary fragments. The data tables by Audi, Wapstra, Thibault [5] were used in the present calculation.

We use in our calculation the level density parameter to be:

$$a(A,Z,U) = a^* \left\{ 1 + \frac{\delta W(A,Z)}{U} \left(1 - e^{-\gamma U} \right) \right\} \tag{7}$$

where $U = E^* - \Delta(A, Z)$, $\gamma = 0.05$, a^* is the asymptotic level density parameter [6]. The pairing Δ and shell correction δW energies for the FF were taken from the nuclear mass formula of Koura et al.[7]. The level density parameters a^* approximate to A/7.25.

The total kinetic energy is used to assess the total FF excitation energy distribution. It is assumed to be approximately Gaussian in shape with an average value and width taken from the experiment (Ref. [3] for spontaneous fission of 252 Cf and Ref. [4] for the neutron induced n(0.53 MeV)+ 235 U reaction).

For sake of simplicity, we have assumed no mass, charge or energy dependence of the cross section for the inverse process of compound nucleus formation. This approximation will be reviewed later on.

We have used the average number of emitted neutrons $\bar{v}(A)$ as a way of partitionning the total excitation energy distribution between the light and heavy fragment. For the spontaneous fission of 252 Cf we used data from Refs. [8, 9]. For the neutron induced $n(0.53 \text{ MeV}) + ^{235}$ U reaction, we used data from Ref. [10, 11].

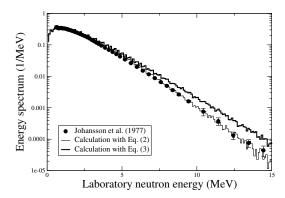


FIGURE 1. Neutron energy spectrum for $n(0.53 \text{ MeV}) + ^{235}\text{U}$ reaction. The thick line is our Monte-Carlo calculation assuming partitioning of FF total excitation energy as a function of $\bar{v}(A)$ and the thin line is the result obtained under the assumption of an equal temperature of complementary FF. The experimental points are from Johansson and Holmqvist [12].

RESULTS AND DISCUSSION

For the neutron-induced reaction on 235 U, the neutron energy spectrum in the laboratory frame is shown in Fig. 1, as calculated using the two different hypotheses for distributing the total available excitation energy among the FF. Also shown for comparison are the experimental data points by Johansson and Holmqvist [12]. The spectrum obtained by assuming equal nuclear temperatures in both FF at scission is shown to agree very well with experimental data, while the alternative hypothesis of splitting the energy according to $\bar{v}_{exp}(A)$ exhibits a much too hard spectrum.

Another physical quantity of interest that can be assessed by our Monte Carlo approach is the neutron multiplicity distribution P(v). Numerical results are compared to the experimental distribution by Diven et al. [13] in Fig. 2. In both calculated cases, the average \bar{v} of the distribution is larger than the experimental value ($\bar{v}_{exp} = 2.47$, Diven et al. [13]). We found $\bar{v} = 2.75$ in the case of equal nuclear temperature for both FF and $\bar{v} = 2.68$ in the other case.

In addition to the multiplicity distribution, the distribution of \bar{v} as a function of A can be inferred from our calculations, and is plotted in Fig. 3. By partitioning the total excitation energy as a function of the experimental values for $\bar{v}(A)_{exp}$ (triangles \triangle are the result obtained under the assumption of an equal temperature of complementary FF, open square symbols \square are obtained for the other assumption), the calculated result is in fair agreement with the data, as expected. On the contrary, the

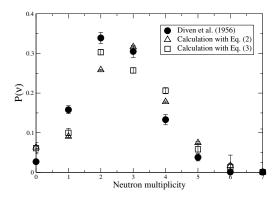


FIGURE 2. Neutron multiplicity distribution for $n(0.53 \text{ MeV}) + ^{235}\text{U}$ reaction . Open square symbols \square are from our Monte-Carlo calculation assuming partitioning of FF total excitation energy as a function of $\bar{v}(A)$, triangles \triangle are the result obtained under the assumption of an equal temperature of complementary FF. The full points are experimental data from Diven et al. [13].

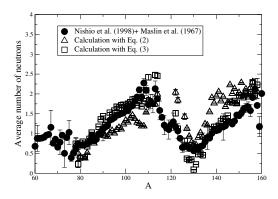


FIGURE 3. Average neutron multiplicity \bar{v} as a function of the mass number of the FF for $n(0.53 \text{ MeV}) + ^{235}\text{U}$ reaction.

equal temperatures assumption is not consistent with the experimental data, although the well-known saw-tooth shape is qualitatively recovered.

In the case of ²⁵²Cf spontaneous fission, similar qualitative conclusions can be drawn, and will therefore not be repeated here, but will be expanded in a longer journal publication. As an example we show the result for the neutron multiplicity distribution in Fig. 4.

We checked the sensitivity of our results upon the various parameters involved in the simulation. It appeared that the limit of the FF excitation energy beyond which no neutrons are emitted is of great importance. In particular, choosing this limit to be equal to the neutron sep-

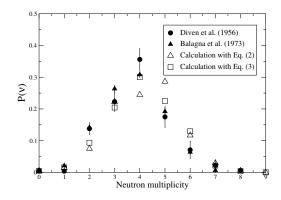


FIGURE 4. Neutron multiplicity distribution for the spontaneous fission of ²⁵²Cf. The full points and triangles are experimental data from Diven et al. [13] and Balagna et al. [14].

aration energy plus pairing energy rather than just the neutron separation energy leads to much better results on neutron energy spectra and neutron multiplicity distributions for both hypotheses of partitioning FF excitation energy. This condition affects our calculation by lowering neutron emission at excitation energy close to the neutron separation energy thus reflecting the increasing competition with gamma ray emissions.

Our calculation is based on a Fermi-gas assumption $E^* = aT^2$. This leads to an overall too high nuclear temperature for low FF excitation energies. An improvement would be to add a constant temperature region to our description of neutron emission sequence for low FF excitation energies and keep the Fermi gas formulation for higher excitation energies. Finally, the cross section for the inverse process of compound nucleus formation will be improved to include a neutron energy dependence.

In conclusion, we have developed a new and powerful tool to explore the process of neutron emission from the statistical decay of FF. The choice of a Monte Carlo implementation to describe this decay process allows to infer important physical quantities that could not be assessed otherwise, for instance within the Los Alamos model framework. In particular, the multiplicity distribution of prompt neutrons P(v), the distribution of v as a function of the FF mass number, and neutron-neutron correlations can all be inferred from the present work.

This simulation tool can also be used to assess the validity of physical input assumptions, in particular the still unanswered question of how does the available total excitation energy get distributed among the light and heavy FF. Further progress of this work will hopefully help to shed some light on this long standing problem.

ACKNOWLEDGMENTS

We are grateful to Dr. F. S. Dietrich and Pr. T. Ohsawa for stimulating and encouraging discussions and Dr. F.-J. Hambsch for providing us with his experimental work.

REFERENCES

- D. G. Madland, J. R. Nix, Nucl. Sci. Eng. 81, 213 (1982).
- 2. V. Weisskopf, Phys. Rev. **52**, 295 (1937).
- 3. F. J. Hambsch, S. Oberstedt, Nucl. Phys. **A617**, 347 (1997)
- H. W. Schmitt, J. H. Neiler, F. J. Walter, Phys. Rev. 141, 1146 (1966).
- G. Audi, A. H. Wapstra, C. Thibault, Nucl. Phys. A729, 337 (2003).
- A. V. Ignatyuk, K. K. Istekov, G. N. Smirenkin, Sov. J. Nucl. Phys., 29, 450 (1979).
- H. Koura, M. Uno, T. Tachibana, M. Yamada, Nucl. Phys., A674, 47 (2000); H. Koura, T. Tachibana, M. Uno, M. Yamada [private communication, 2004].
- 8. R. L. Walsh, J. W. Boldeman, Nucl. Phys. **A276**, 189 (1977).
- C. Budtz-Jørgensen, H. H. Knitter, Nucl. Phys. A490, 307 (1988).
- K. Nishio, Y. Nakagome, H. Yamamoto, I. Kimura, Nucl. Phys. A632, 540 (1998).
- E. E. Maslin, A. L. Rodgers, W. G. F. Core, Phys. Rev. 164, 1520 (1967).
- P. I. Johansson and B. Holmqvist, Nucl. Sci. Eng. 62, 695 (1977).
- B. C. Diven, H. C. Martin, R. F. Taschek and J. Terrell, Phys. Rev. 101, 1012 (1956).
- J. P. Balagna, J. A. Farrell, G. P. Ford, A. Hemmendinger, D. C. Hoffmann, L. R. Vesser and J. B. Wilhelmy, in Proceedings of the Third International Atomic Energy Symposium on the Physics and Chemistry of fission, Rochester, 1973, Vol. 2, p. 191 (IAEA, Vienna, 1974).